

Supporting Information

Ligand-Mediated, Temperature-Tuned Synthesis of CsPbBr₃ Nanosheets for Ordered Superlattice Assembly

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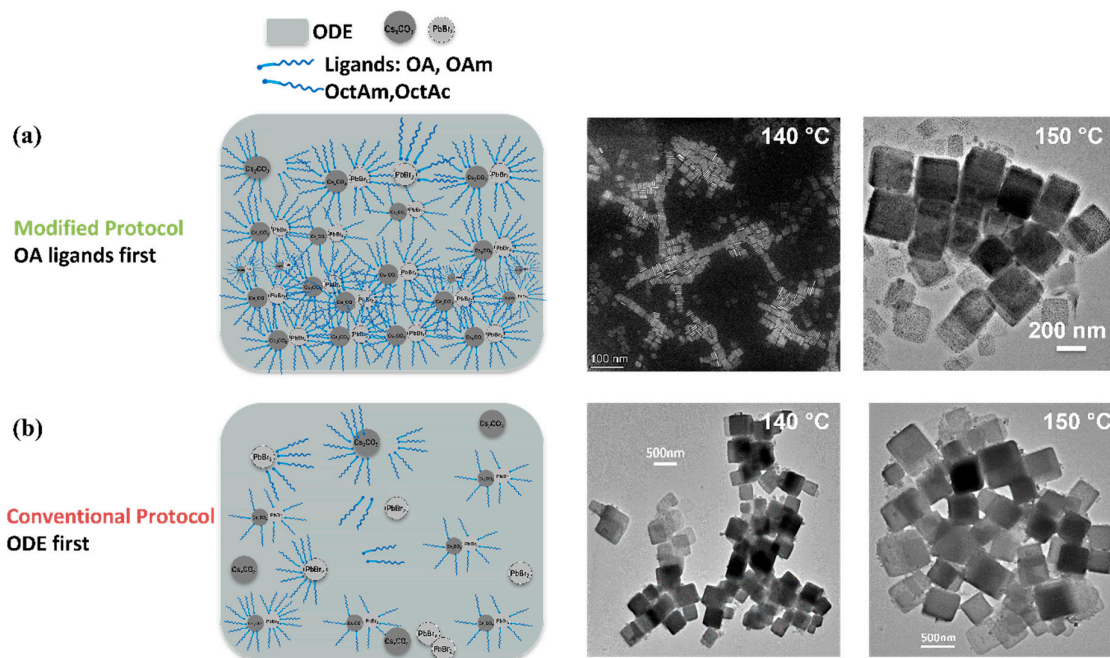


Figure S1. (a) Schematic illustration of ligand–precursor interactions in the modified protocol, where oleic acid (OA) is introduced and mixed with the precursor prior to the addition of the solvent 1-octadecene (ODE). In this approach, OA ligands bind effectively to the precursors, promoting controlled crystal growth and facilitating the self-assembly of the nanosheets. Representative TEM images of nanosheets synthesized at 140 °C and 150 °C using this method are shown on the right. (b) Schematic diagram of the conventional hot-injection synthesis, in which ODE is added first, followed by the precursors and ligands. In this case,

OA interacts weakly with the precursors during the initial reaction stage, resulting in less regulated crystal growth and partial ligand loss during purification. The corresponding TEM images (right) reveal poor growth control and reduced nanosheet self-assembly into superlattices

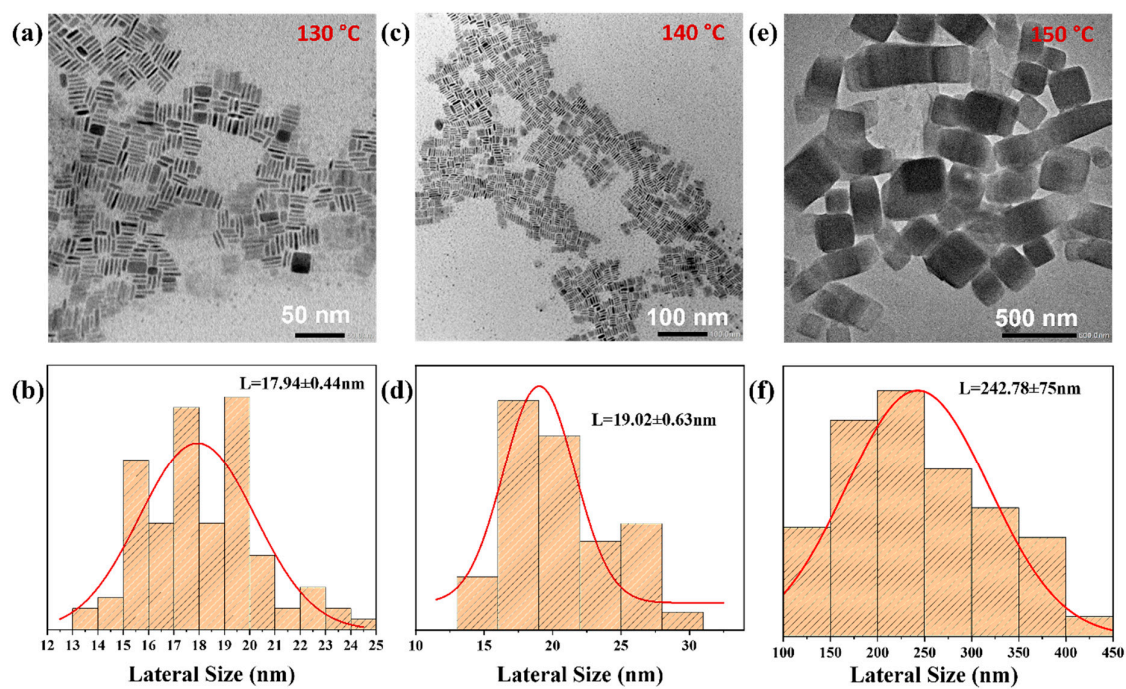


Figure S2. TEM images (top) and corresponding lateral size distributions (bottom) of 2D CsPbBr₃ nanosheet superlattices synthesized at different temperatures: (a,b) 130 °C, (c,d) 140 °C, and (e,f) 150 °C

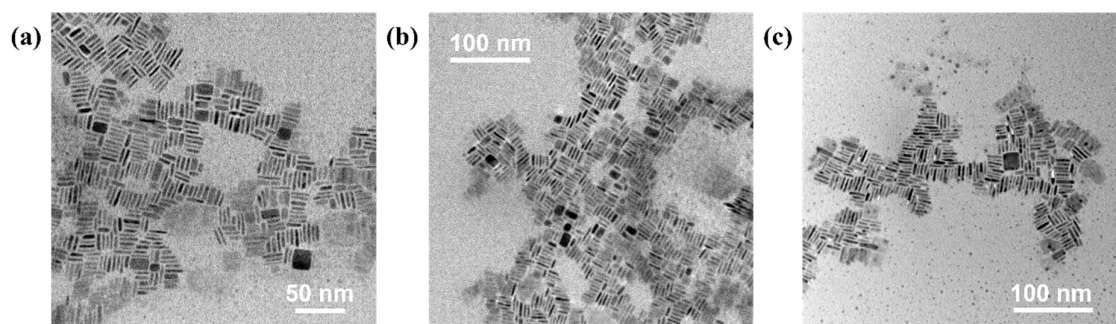


Figure S3. (a-c) TEM image of 2D CsPbBr₃ NSs superlattice at to 130 °C

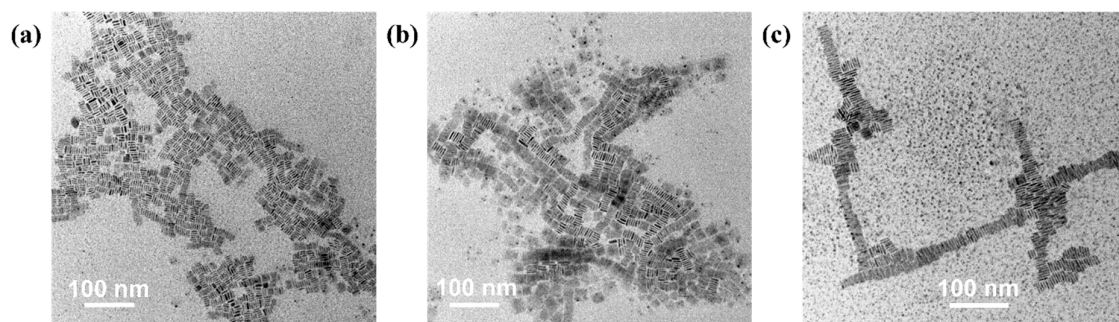


Figure S4. (a-c) TEM image of 2D CsPbBr₃ NSs superlattice at to 140 °C

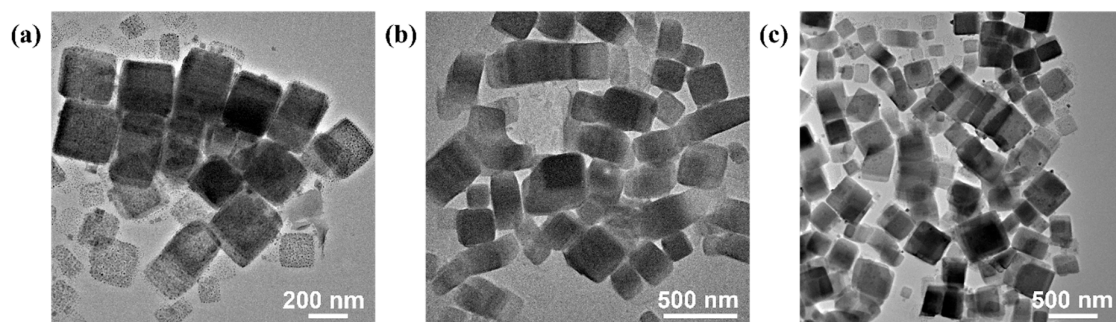


Figure S5. (a-c) TEM image of 2D CsPbBr₃ NSs superlattice at to 150 °C

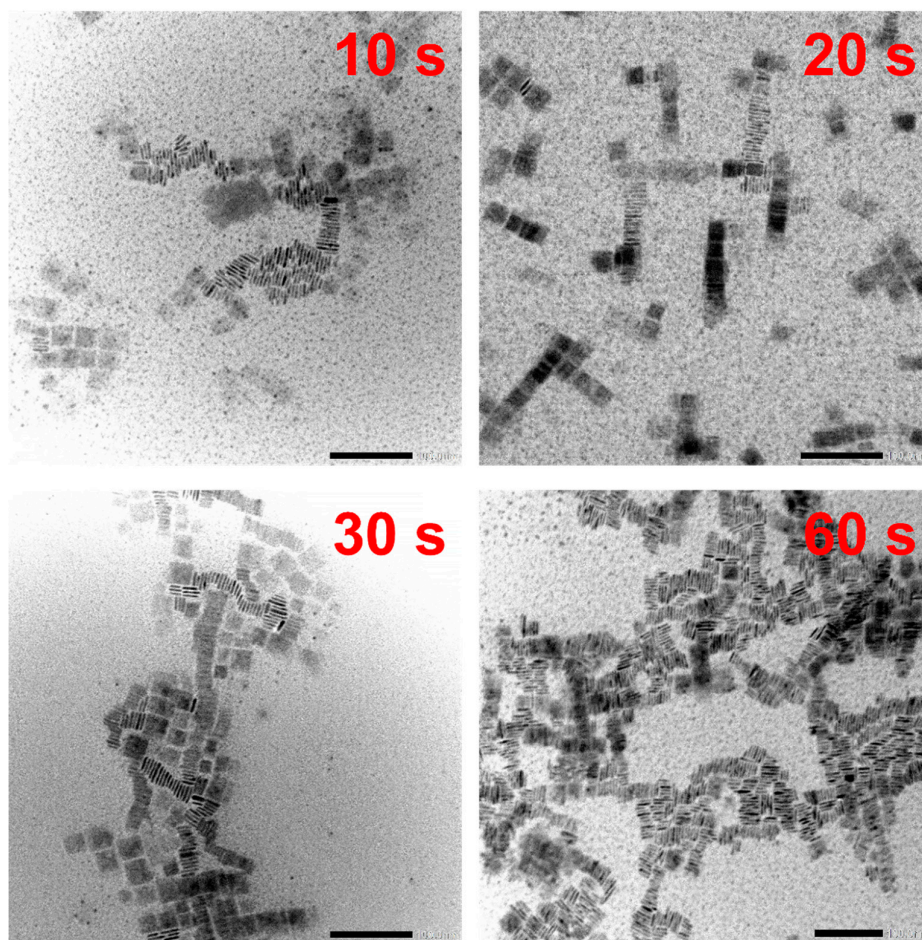


Figure S6. TEM image of 2D CsPbBr₃ nanosheet superlattices showing the morphology and self-assembly of nanosheets at different reaction times, highlighting the size evolution and formation of ordered superstructures

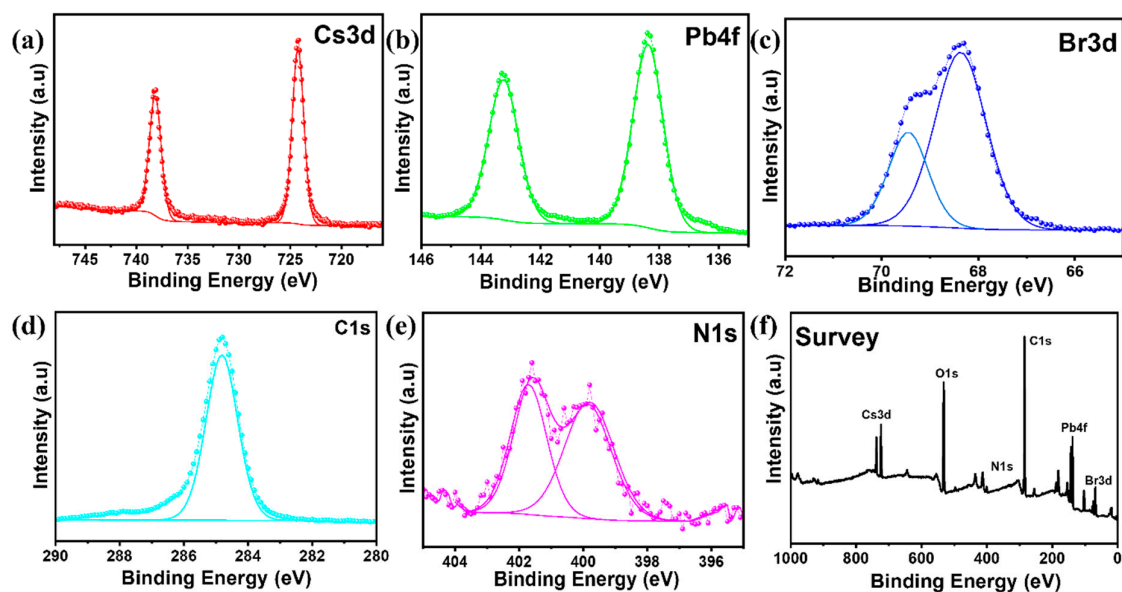


Figure S7. (a-f) XPS profile of CsPbBr₃ NSs superlattice (a) Cs3d, (b) Pb4f, (c) Br3d, (d) C1s, (e) N1s, (f) Survey, elements for CsPbBr₃ nanosheets superlattice.

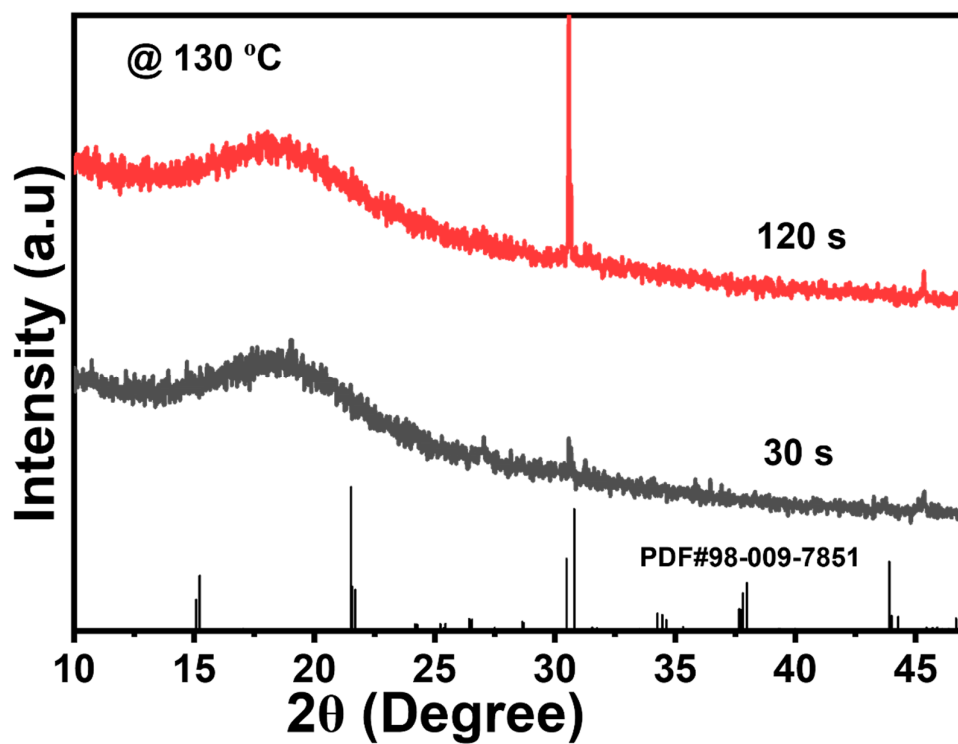


Figure S8. XRD patterns of CsPbBr₃ nanosheets synthesized at 130 °C with reaction times of 30 s and 120 s. The 30 s sample exhibits a broad, featureless background indicative of amorphous Pb-Br-rich residues, while the 120 s sample shows a weak emerging diffraction peak alongside the amorphous halo, suggesting the onset of CsPbBr₃ crystallization under prolonged reaction time.

Table S1. Atomic composition of CsPbBr₃ determined by XPS.

Elements	Atomic % (XPS)	Atomic Ratio
Cs3d	20.18	1.00
Pb4f	19.92	0.99
Br3d	68.45	3.39
C1s	2.07	-
N1s	1.38	-

Table S2. Detailed recombination lifetimes determined using an exponential fitting approach.

Sample	Γ_1 (ns)	A ₁ (%)	Γ_2 (ns)	A ₂ (%)	Γ_3 (ns)	A ₃ (%)	Γ_{ave} (ns)
CsPbBr ₃ @130	2.75	37.91	8.01	53.26	37.90	8.82	8.65
CsPbBr ₃ @140	11.47	89.29	48.38	10.71	-	-	15.42
CsPbBr ₃ @150	1.24	20.28	9.95	40.32	79.25	39.40	35.49

The time-resolved PL decay curves were fitted using a multi-exponential function:

$$Y = A_1 \exp\left(-t/\Gamma_1\right) + A_2 \exp\left(-t/\Gamma_2\right) + A_3 \exp\left(-t/\Gamma_3\right)$$

The average lifetime Γ_{ave} was calculated according to:

$$\Gamma_{ave} = \sum A_i \Gamma_i^2 / \sum A_i \Gamma_i$$

Where, Γ_1 represents the short-lived component, typically attributed to radiative recombination, while the longer-lived terms (Γ_2 for 140 °C and Γ_3 for 130 °C and 150 °C) are mainly associated with non-radiative processes. The coefficients A₁, A₂ and A₃ denote the relative weighting of each recombination pathway in the overall decay profile.

Table S3. Detailed synthesis temperature, Lateral size, Absorption, Photoluminescence and Stoke Shift of nanosheets superlattice.

Sample	Lateral size (nm)	Absorption (Abs, nm)	Photoluminescence (PL, nm)	Stoke Shift (nm)	Exciton/Trap State
CsPbBr ₃ @130	17.9 ± 0.44	457	462	5	Free or weakly bound exciton
CsPbBr ₃ @140	19.02 ± 0.63	458	464	6	Weakly/ Intermediate trapping
CsPbBr ₃ @150	242.78 ± 75	479.16	513	33.84	Self-trapped exciton